Workshop on Detectors for Synchrotron Research

Recommendations of the Diffraction & Scattering working group.

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Executive summary
X-ray diffraction and scattering encompasses an enormous diversity of experimental tools and techniques, as applied to practically every material imaginable. Examples include crystallography, scattering from poorly ordered materials, solution scattering, small angle x-ray scattering (SAXS), grazing incidence x-ray scattering (GIXS), scattering from surfaces, etc. Not surprisingly, performance of these experiments requires a corresponding diversity of x-ray detector tools. Examination of one case example after another indicates, however, that the major instrumental limitations arise not from the x-ray sources or the beamlines but from the absence of detectors capable of capturing all the data made possible by advanced synchrotron sources.

The ideal detector would extract the maximum possible information from each scattered x-ray photon, that is the exact position (x,y), time of arrival (t), wavelength (λ), angle of incidence (x',y'), polarization (P), and phase (ϕ) of each x-ray over a detector surface of defined figure (e.g. planar, cylindrical, spherical). No detector in existence records even half of these parameters, and even then, detection is always over a limited range and with limited accuracy. Recent developments in integrated circuit, materials, and superconducting technology suggest that, given adequate R&D, new detectors are feasible which would be superior to any now available, and which would dramatically enable new science.

Below, we list some examples of important detector-limited experiments and comment on detectors which might be developed to meet the experimental needs. It is to be understood that these are merely suggestions and that more comprehensive investigation might reveal other detector technologies which might be more feasible to develop for the given application. We conclude with general recommendations.

Combined X-ray Crystallography and Holography
Miao and Sayre (refs) have suggested that oversampling of x-ray crystallographic patterns may be used to extract phase information and, thereby, to circumvent to phase problem in crystallography. The phase problem, as is well known, is the major hurdle which must be overcome in solving crystallographic structures and accounts for much of the effort of crystallographers. Even if the phase problem is not completely resolved by oversampling techniques, the additional constraints imposed by combining oversampling with other methods will assist in the structure refinement process. Oversampling may be accomplished by simultaneously recording Bragg peaks and the diffuse, continuous scatter between the peaks. Since the peaks are many orders of magnitude brighter than the inter-peak scatter, the constraints on the detector are that the Point Spread Function (PSF) fall by 4-6 orders of magnitude in a distance small compared to the inter-Bragg peak spacing. Wall et al. ( ) used a CCD detector with state-of-the-art resolution to perform such an experiment on a protein crystal and found that much of the data had to be discarded because of low-level wings of the PSF arising from light spreading in the phosphor and fiber optics. PSF limitations would be even

more severe with image plate detectors. Moreover, the very high count rates from many patterns, when combined with the very large numbers of x-rays required for accurate measurements and the need to use samples efficiently to limit radiation damage, render inadequate all existing alternative detectors. The low level PSF results from physical processes (e.g., light scatter) which are absent in the direct x-ray to electron-hole conversion process used by, for example, Pixel Array Detectors (PADs) and active matrix pixel detectors. Indeed recent measurements on the PSF of PADs, in agreement with theory (Janesick, ???), show no detectable signal spread beyond nearest neighboring pixels. This is an example of how practical large-area pixel detectors, were they to be available, would open up whole new areas of science.

Laue Diffraction

Monochromatic x-ray crystallography is limited by the need to rotate the crystal in order to pass the reciprocal lattice points through the Ewald sphere. In Laue crystallography, a polychromatic beam provides a thick Ewald sphere wall which inherently encompasses a large volume of reciprocal space so that many Bragg spots are simultaneously reflecting. In principle, this allows time-resolved crystallography down to extraordinarily short times. Moffat and coworkers (ref) have explored the possibilities presented by time-resolved Laue diffraction of protein crystals. The Laue method is found to be limited by crystal mosaicity and detectors. Thermally-induced mosaicity most commonly arises from heat accompanying, for example, optical laser triggering of a chromaphore-induced event in a protein. This is often a detector problem in disguise, because the increase in mosaicity may arise on much slower time scales than many of the interesting changes in the protein. If it were possible to acquire several full Laue images in succession on microsecond or submicrosecond time scales, then it might be possible to obtain the requisite diffraction patterns before crystal heating is manifest. Another limitation of Laue diffraction is the need to isolate the diffraction patterns on times faster than achievable with x-ray shutters. This has usually required operation of storage rings in asymmetric bunch patterns in which an isolated bunch or bunch train is used to define a time interval long enough to allow shuttering of the experiment. This unusual mode of storage ring operation impacts other users and, in consequence, can only be performed at very specially scheduled times. Again, the absence of suitable detectors which can be electronically shuttered in the requisite time frames, is limiting the progress of Laue crystallography.

What is needed is a detector which can record a sequence of Laue diffraction patterns in successive, electronically synchronized and programmable microsecond or submicrosecond time intervals so as to allow the use of single crystals without special storage ring modes. It is entirely feasible to do this, at least down to a few hundred nanoseconds, with an analog PAD. Thinking even more broadly, some of the Laue reflections contain several x-ray wavelengths. The ideal would be to be able to record the Laue patterns in such a way as to reject x-rays below a given wavelength. This is a very ambitious goal, but one which is certainly suitable for long-range detector research.

Time-Resolved Oscillatory Materials

Many materials undergo oscillatory structural changes, or can be made to do so in synchrony with a periodic signal. Examples include stretch-activated insect flight muscle, propagating

phonon modes, oscillatory elastically strained or heated materials, liquid crystal reorientation in RF fields, etc. In many of these cases, the x-ray diffraction differences of interest during different parts of the oscillatory cycle are very small and the acquisition of accurate data requires signal averaging over many cycles. Many samples exhibit slow fatigue, drift, radiation damage or large sample-to-sample variations (e.g., due to absorption) which complicate or limit the ability to integrate low noise diffraction from different parts of the oscillatory cycle or from multiple samples for later subtraction to uncover the small fractional differences. In other cases, the frequency of oscillation is simply too high for ready isolation of a small part of the oscillatory diffraction.

For single point signals, the time-honored way to deal with such low-level signals is to use phase sensitive lock-in amplifier methods. It is entirely feasible to develop x-ray detectors in which each pixel over a 2-dimensional surface effectively acts as a lock-in amplifier. For example, PADs can be designed in which the signal from each pixel is successively either added into or subtracted from an in-pixel integrating capacitor in synchrony with an external electronic clocking signal. It is even possible to put several integrating capacitors into each pixel to divide the oscillatory cycle into small parts (e.g. see Rossi et al (199?). Such analog PADs can have effectively long integration times to extract very small differences over an enormous number of cycles, even if the count rates are very high. Alternatively, for lower count rates (MHz /pix) digital PADs can directly bin the counts in-pixel into externally phase-locked bins with depths of, say, 32 bits. The availability of such point-by-point area lock-in x-ray detectors would substantially expand the feasibility of many kinds of very low-level difference experiments.

Photon Counting Diffractometry

The frontier of macromolecular crystallography involves the determination of atomic structure in cases where the inherent signal contrast (e.g., signal-to-background ratio) is very low. Examples with very low contrast include poorly ordered crystals, crystals with very high water content, very large unit cell macromolecular assemblies, and all crystals at the limits of their observable diffraction resolution. Simply increasing the exposure time is not an option in many cases because the crystals are already at radiation damage limits; indeed, radiation damage to a given contrast level and resolution set the smallest crystal size which can be practically utilized. Since small crystals are more easily obtained than larger ones, signal contrast limitations are the major limitation on the number of structures which are feasible.

Ideally, in monochromatic oscillation crystallography one wishes to record the diffraction from a given Bragg reflection only when the corresponding reciprocal lattice point is passing through the Ewald sphere, i.e., when it is in a diffracting condition. The optimum signal-to-noise ratio is then achieved by subtracting the time-normalized background signal immediately before and after the spot passes through the Ewald sphere. This procedure automatically takes into account slowly changing background levels and helps to remove detector flat-field variations. This is the essence of the so-called phi- or fine-slicing method. By contrast, CCD or image plate detectors operate in integrate/readout cycles which, in practical terms, means that the user selects an oscillation range and records all reflections over that range. Since different spots pass through the Ewald sphere at different times, any fixed oscillation range will necessarily involve the recording of reflections which are only partially through the Ewald sphere at the ends of the

oscillation range. The scaling and merging of these partials often limit the quality of the data, so the user is forced to compromise between a wider oscillation range, which reduces the fraction of partials, and a fine oscillation range, which can approach more ideal fine-slicing. A better approach would be to develop PAD or other photon counting detectors which record the data into user-selectable, arbitrarily short time-bins which may vary asynchronously across the face of the detector. Such detectors do not now exist, at least at the requisite count-rates, but are clearly possible.

Micro-Resolution Imaging for Hard X-rays

X-ray microtomography allows the 3-dimensional reconstruction of minerals, electronic nanostructures, fiber optical components etc. Topography is another powerful technique which is useful for visualization of crystal defects. These methods will become of increasing importance as electronic and mechanical structures shrink deeper into the submicron regime. Metrology of small structures must necessarily be an important part of the national nanoscale initiative. Another important application is in the determination the 3-dimensional structure of mineral inclusions, grains, and microcrystals. The application of these methods to specimens containing high atomic weight atoms, or fabricated on thick substrates, usually requires hard (> 15 keV) x-rays to mitigate specimen absorption effects. In these cases, tomographic and topographic methods are limited by the lack of electronic imaging detectors capable of efficiently recording hard x-ray patterns with submicron resolution. The alternatives, namely scanning microbeam methods are often too inefficient for an acceptable data flow. The development of detectors with very high spatial resolution and good efficiency at short x-ray wavelengths would considerably expand the kinds of experiments which would be performed.

The present limit of area detectors is about ??? microns (ref ESRF work), which is already inadequate for many specimens of interest. There is a strong need to develop higher spatial resolution hard x-ray imaging detectors. For example, it may be possible to fabricate a close-packed array of long, very high atomic weight, electronically isolated sub-micron diameter columns of reverse-biased semiconductor materials which act as radiation sensitive diodes and which provide adequate stopping power perpendicular to the detector face, but limit the in-plane signal spread. Another possibility might be to fabricate an array of free-standing metal fibers, each connected to a superconducting bolometer detector. If such a detector with high count-rates could be fabricated, then the sharp energy resolution of the bolometers would also allow innovative experiments, such as tomography with simultaneous elemental analysis by using a white or a pink beam which spans the K-edge of desired elements. Micro tomography and topography are examples where there is a clear detector need requiring some innovative long-range thinking.

X-ray Photon Correlation Spectroscopy (PCS)

X-ray PCS is an emerging technique to examine the dynamics of materials at length scales down to the nanometer with time resolutions in the 10^{-3} to 10^{-9} range (ref). Examples of materials which may be examined via x-ray PCS include the inherent dynamics and relaxation dynamics of colloids, polymers, liquid crystals, glasses, solid-state phase transitions, and homogeneous solid state chemical reactions. PCS relies on the time autocorrelation of scattered x-ray speckles, in

exactly the same way as optical PCS: the movement of scattering centers in the sample causes slight shifts in the phase of the scattered rays which dynamically vary the net constructive or destructive interference pattern on a distant detector surface. The size of the speckles encountered at, for example, the APS or the ESRF tend to be in range of a few to a few tens of microns with sample to detector distances of up to a few meters. This size range is smaller than the pixels of most CCD or image plate detectors. Also, PCS patterns are dynamic entities for which the readout per pixel must be on the relevant time scale to extract the desired autocorrelation function. The slow readout of existing CCD detectors is limiting.

The immediate need is for detectors which have resolution on the order of 5-10 microns, a wide dynamic range, photon limited sensitivity and very rapid readout. Custom deep depletion CCDs with many readout ports are possible and would serve this application well. An alternative might be the active matrix pixel detectors suggested by the Pavel Rehak at BNL. In both of these cases, however, readout times for large format area imagers is probably limited to about a millisecond. Longer range alternatives suitable for shorter time scales involve PADs with in-pixel autocorrelators, which are entirely feasible with small scale integrated circuit integration for pixels in the 50 - 100 micron range. The larger pixels will necessitate longer distances between the specimen and the detector, but such distances are feasible on many beamlines.

Wide Solid Angle Powder Diffractometry and Fluorescent Energy Analysis

Even some of the most fundamental experiments are limited by available detectors. Two examples are powder diffractometry and elemental distributions by analysis of the x-ray fluorescence. In powder diffractometry, the 1-dimensional x-ray diffraction from a randomly oriented powder of a material is recorded and analyzed (ref). Synchrotron radiation makes possible the recording of a very large number of Debye-Scherrer rings, which provide sufficient information to determine the structure of surprisingly large unit cells, perhaps even as large as proteins (e.g., ref). The primary advantage of the powder technique is the absence of a need to obtain single large crystals. However, in order to maximize the information obtained per unit dose of incident radiation, it is necessary to record the diffraction to very large angles, frequently as large as $2\theta \quad 180°$, i.e., nearly fully back-scattered. The challenge, then is to devise simple detectors which view the diffraction over as much of 4π steradians as possible. This is inherently a 1-dimensional problems, although the goal is to cover as much of an enclosing surface as possible.

Another fundamental application is elemental analysis by energy analyzing the x-ray fluorescence off a sample. This application is especially powerful if a micro x-ray beam in used to scan the sample. Again, the challenge is to combine very good energy resolution with as much solid angle coverage as possible. In this case, however, 2π steradian coverage is typically ideal.

These applications are presently limited by the count-rate, energy resolution, and solid angle of coverage of available detectors. It is feasible to devise a variety of detector solutions based on PADs, active matrix pixel detectors or silicon drift detectors. In the interests of efficiency, the detector elements should be arranged in the form of tiles which cover a large area, perhaps in the form of a “bucky ball”. The availability of efficient, large solid angle detectors for these two applications would greatly increase the number of samples which may be analyzed and advance

the limits now imposed by specimen size, elemental concentration, unit cell size or radiation damage lifetime.

General Recommendations

1. *The best currently available detector technology needs to be installed on beamlines.* In a surprising number of cases, very expensive beamlines are limited by the inability to procure the best currently available detectors. This situation is especially true for beamlines which do not specialize on biological macromolecular crystallography. It makes no sense to limit beamline capabilities for the lack of funds for detectors costing much less than the cost of the beamlines, and much, much less than the cost of the synchrotron ring.

2. *The funding agencies should actively promote better cooperation between detector groups in the U.S. and abroad.* The Europeans, especially, are making great advances in many pixel detector technologies. More cooperation and collaboration would benefit everyone involved, especially since the funding resources come from distinct pools. Cooperation may take many forms: joint workshops, visitor exchange programs, web publication of details of detector advances not suitable for journal publication, and cooperative agreements which allow access to integrated circuit libraries and software.

3. *Mechanisms are needed to allow acquisition of advanced detectors for which the market is too small to attract industry*. The explosion of research based on biomacromolecular crystallography was enabled by the availability of image plate and CCD detectors. In this case, the market for a single kind of diffraction experiment was large enough to attract industry, with the result that a steady stream of incremental advances are now proceeding with little need for government intervention. However, few of the diverse applications of encompassed by diffraction and scattering techniques are able to boast such a market; rather, the needs are large, but the specific methods are diverse. Detectors suitable for many of these needs are often developed by individual detector groups, but these groups are ill-suited to provide more than a few detectors. In the absence of industrial participation, there is no effective mechanism to make these detector advances available to the larger community.

   Innovative mechanisms are needed to solve this problem. An appropriate solution might be to utilize SBIR funds for this purpose. Most commonly, SBIR funds are now used to encourage industry to perform the basic detector research. An alternative might be to identify detector advances which have already been demonstrated and specifically issue an SBIR competition for duplication and dissemination, with appropriate care for compensation to the original developers of the technology. This has many advantages: It opens a pool of funding already allocated, it provides a relatively low risk inducement for industry, and it is a mechanism for disseminating the fruits of small detector groups.

4. *The number and size of detector groups in the U.S. needs to be dramatically increased, both at the synchrotron sources and in the universities.* The ESRF and Daresbury detector groups are each larger than the combined detector groups at all of the major U.S.

hard x-ray synchrotron sources (NSLS, APS, ALS, SSRL, CAMD and CHESS) combined! The absence of in-house detector groups at the sources will limit the effective utilization of detector technology, even if this technology were to become available, because detector expertise is required to integrate state-of-the-art detectors into experiments. Due to lack of funds, detector groups, both at the sources and elsewhere have shrunk both in size and number. An infusion of funds is desperately needed to reverse this situation. There is need both for groups to help users with existing detectors at the sources and, most especially, for groups to develop new detectors. Detector advances have historically come from many different sectors: Universities, national labs and industry. Therefore, it is important that funds for detector development be made available based on competitive merit of the proposals, and not simply by the location or sector. The criteria for funding the proposals must be based solely on a wise combination of factors of (a) a demonstrated record of detector accomplishments, (b) users needs, and (c) the innovation and promise of the proposed detector research.

5. *Detector research needs to balance shorter term needs with long term basic research.* Incremental improvements on, for example, high energy or high resolution CCD phosphors, would have an enormous impact on the utilization of existing CCD detector technology. These are likely to be short-term projects. On the other hand, very promising future technologies, such as Pixel Array Detectors (PADs), active matrix pixel detectors, superconducting detectors, etc. will require gestation periods of a decade or more. There is a need to fund both short-term application needs and more risky longer term research.

6. *A standing interagency standing detector committee should be formed to identify opportunities and to suggest ways in which detector development might advance.* Detector developments take a very long time – frequently decades -- before reaching fruition. While a Detector Initiative would be welcome, especially in terms of providing existing technology, sustained long-term support is required if detector advances are to continue. A standing interagency committee should be formed to advise the agencies of detector areas in need of attention. This advice might include things like suggestions of detector opportunities for Requests For Proposals (RFPs), suggestions for workshops and exchange programs to promote international cooperation, and suggestions for leveraging synergy with other communities with related technological problems (e.g., medical radiology groups, or the high energy physics community).